

IN THE CLAIMS

A complete listing of the pending claims follows:

1. (currently amended): A method of in-process ratio automated isotope dilution mass spectrometry comprising

providing a sample,

providing a spike ~~spiking at least one enriched stable isotope of an element or specie~~ related to said sample,

~~introducing said spiked enriched stable isotope elements or species into said sample~~
spiking the sample with the spike and permitting equilibrium to occur therebetween,

subjecting said equilibrated spike spikes and sample to atmospheric pressure ionization to create ions therefrom,

introducing said ions into a mass spectrometer for a isotopic ratio determination, and
~~delivering information from said determination to a microprocessor~~

in a processor, using the ratio determination to characterize the sample.

2. (currently amended): The method of in-process ratio automated isotope dilution mass spectrometry of claim 1 including wherein the act of providing a sample comprises providing a liquid sample ~~employing a liquid sample as said sample.~~

3. (currently amended): The method of in-process ratio automated isotope dilution mass spectrometry of claim 2 including wherein the act of providing a liquid sample comprises providing an aqueous sample ~~employing an aqueous solution as said sample.~~

Claims 4 – 5 (cancelled):

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6. (original): The method of in-process ratio automated isotope dilution mass spectrometry of claim 1 wherein the act of providing a sample comprises providing a sample having one or more contaminants including employing said process to detect levels of one or more contaminants in a sample.

7. (currently amended): The method of in-process ratio automated isotope dilution mass spectrometry of claim 6 including
detecting said contaminants at near instrument detection limits.

8. (currently amended): The method of in-process ratio automated isotope dilution mass spectrometry of claim 6 including
detecting said contaminants at ultra-trace levels.

9. (currently amended): The method of in-process ratio automated isotope dilution mass spectrometry of claim 1 including
after said equilibration but before said ionization, preconcentrating the spike and sample said elements or species.

10. (currently amended): The method of in-process ratio automated isotope dilution mass spectrometry of claim 9 including
effecting said preconcentration through liquid chromatography.

11. (currently amended): The method of in-process ratio automated isotope dilution mass spectrometry of claim 9 including

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separating at least one ~~specie~~ species of interest by said preconcentration.

12. (currently amended) The method of in-process ratio ~~automated isotope dilution~~ mass spectrometry of claim 1 ~~including~~ further comprising
employing said method in qualitative analysis of an analyte in the sample of said
~~element or species.~~

13. (currently amended) The method of in-process ratio ~~automated isotope dilution~~ mass spectrometry of claim 1 ~~including~~ further comprising
employing said method in quantitative analysis of an analyte in the sample of said
~~element or species.~~

14. (currently amended) The method of in-process ratio ~~automated isotope dilution~~ mass spectrometry of claim 1 including
employing information received by said ~~microprocessor~~ processor to control operation
of portions of said method.

15. (currently amended) The method of in-process ratio ~~automated isotope dilution~~ mass spectrometry of claim 1 ~~including~~ further comprising
obtaining said sample from a system being monitored, and
delivering ~~a portion of~~ the information received by said ~~microprocessor~~ processor
regarding the characterization of the sample to said system from which the sample was
obtained.

16. (currently amended): The method of in-process ratio ~~automated isotope dilution~~

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mass spectrometry of claim 1 including

employing said method to monitor concentration of analytes ~~contaminants in a plurality of wet baths employed in clean rooms~~ in semiconductor manufacture.

17. (currently amended): The method of in-process ratio ~~automated isotope dilution~~ mass spectrometry of claim 16 including

employing said method sequentially on a plurality of said wet baths used in the semiconductor manufacturing.

18. (currently amended): The method of in-process ratio ~~automated isotope dilution~~ mass spectrometry of claim 16 including

employing said method simultaneously on a plurality of said wet baths used in the semiconductor manufacturing.

19. (currently amended): The method of in-process ratio ~~automated isotope dilution~~ mass spectrometry of claim 1 wherein the act of providing the sample comprises providing a gaseous sample ~~including employing a gaseous specimen as said sample.~~

20. (currently amended): The method of in-process ratio ~~automated isotope dilution~~ mass spectrometry of claim 1 including

employing electrospray ionization as said atmospheric pressure ionization.

Claims 21 through 22 (cancelled).

23. (currently amended): The method of in-process ratio ~~automated isotope dilution~~

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mass spectrometry of claim 16 including

employing said method to determine ~~which said bath was the origin of the analytes~~
~~said species or elements.~~

24. (currently amended): ~~The A~~ method of automated isotope dilution mass
spectrometry ~~of claim 1 including~~ comprising

providing a sample to be analyzed,

spiking at least one enriched stable isotope of an element or specie related to said
sample,

introducing said spiked enriched stable isotope elements or species into said sample
and permitting equilibrium to occur therebetween,

subjecting said equilibrated spikes and sample to atmospheric pressure ionization to
create ions therefrom,

introducing said ions into a mass spectrometer for isotopic ratio determination,

delivering information from said determination to a microprocessor, and

in effecting said equilibrium equilibrating at least one said spiked enriched stable
isotopic specie or element dynamically with a specie or element contained within the
sample.

25. (currently amended): Apparatus for in-process ratio ~~automated isotope dilution~~
mass spectrometry comprising

sample receiving apparatus adapted to receive a sample,

spike introduction apparatus for introducing at least one isotopically enriched spike
~~spiked enriched stable isotope element or specie~~ into said sample for permitting equilibration
therebetween,

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an atmospheric pressure ionizer for receiving said equilibrated sample and spike
~~spiked elements or species~~ and ionizing the same,

a mass spectrometer for receiving and processing said ions by isotope ratio
determination, and

a processor adapted to use the ratio to characterize the sample ~~microprocessor for~~
~~receiving information about said determination from said mass spectrometer.~~

Claim 26 (currently amended): The in-process ratio ~~automated isotope dilution~~ mass
spectrometry apparatus of claim 25 including further comprising

a sample analyzer for analyzing said sample and delivering sample analysis
information to said processor ~~microprocessor~~.

27. (currently amended): The in-process ratio ~~automated isotope dilution~~ mass
spectrometry apparatus of claim 26 including further comprising

a controller for receiving information processed by said ~~microprocessor~~ processor and
providing feedback to other portions of said apparatus.

28. (currently amended): The in-process ratio ~~automated isotope dilution~~ mass
spectrometry apparatus of claim 25 including further comprising

a controller for receiving information processed by said ~~microprocessor~~ processor and
providing feedback to other portions of said apparatus, and

sample modification apparatus for altering characteristics of said sample responsive to
signals from said controller prior to the sample entering said atmospheric pressure ionizer.

29. (currently amended): The in-process ratio ~~automated isotope dilution~~ mass

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spectrometry apparatus of claim 28 ~~including~~ wherein

said controller is configured to coordinate ~~coordinating~~ operation of said sample receiving apparatus, said spike introduction apparatus, said sample modification apparatus, said atmospheric ion generator and said mass spectrometer.

30. (currently amended): The in-process ratio ~~automated isotope dilution~~ mass spectrometry apparatus of claim 29 ~~including~~ further comprising

solution handling apparatus interposed between said sample modification apparatus and said atmospheric ion generator, and

at least one chromatograph operatively associated with said solution handling unit for preconcentrating said equilibrated sample and spike ~~spiked-enriched stable isotope elements or species~~ prior to delivery to said atmospheric ion generator.

31. (currently amended): The in-process ratio ~~automated isotope dilution~~ mass spectrometry apparatus of claim 30 ~~including~~ wherein

said chromatograph ~~including at least one chromatograph~~ is selected from the group consisting of a liquid chromatograph and a gas chromatograph.

32. (currently amended): The in-process ratio ~~automated isotope dilution~~ mass spectrometry apparatus of claim 30 ~~including~~ wherein

said sample receiving apparatus includes ~~having~~ a first outlet conduit in communication with said spike introduction apparatus which in turn has an outlet conduit in communication with said ~~chemical~~ sample modification apparatus and a second conduit in communication with said ~~chemical~~ sample modification apparatus ~~whereby a sample may be~~ admixed with said spiked-enriched stable isotope elements or species in effecting

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~~quantitative analysis and may be introduced directly into said chemical modification apparatus without admixture with said spiked elements or species for effecting qualitative analysis.~~

33. (currently amended): The in-process ratio ~~automated isotope dilution~~ mass spectrometry apparatus of claim 25 ~~including~~ wherein

said atmospheric ion generator ~~being~~ is an electrospray ionizer.

34. (currently amended): The in-process ratio ~~automated isotope dilution~~ mass spectrometry apparatus of claim 25 ~~including~~ wherein

said atmospheric ion generator ~~being~~ is structured to operate at a first voltage when ~~effecting ionization of multiple enriched stable isotopic elements~~ the apparatus is characterizing an element in the sample and a lower second voltage when ~~ionizing multiple enriched stable isotope species~~ when the apparatus is characterizing a species in the sample.

35. (currently amended): The in-process ratio ~~automated isotope dilution~~ mass spectrometry apparatus of claim 34 ~~including~~ wherein

said first voltage ~~being~~ is 200 to 1,000 volts and said second voltage ~~being~~ is 2 to 30 volts.

36. (currently amended): The in-process ratio ~~automated isotope dilution~~ mass spectrometry apparatus of claim 25 ~~including~~ further comprising

a system interface for receiving information from said microprocessor processor and providing feedback to the system being monitored.

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37. (currently amended): The in-process ratio ~~automated isotope dilution~~ mass spectrometry apparatus of claim 36 including wherein

said system interface having includes a warning capability if the concentration of a monitored contaminant approaches a tolerable ~~upper~~ limit thereof and an alarm capability if the concentration of said contaminant violates ~~reaches or exceeds~~ the tolerable ~~upper~~ limit.

38. (currently amended): The in-process ratio ~~automated isotope dilution~~ mass spectrometry apparatus of claim 25 including wherein

said atmospheric ion generator ~~being is~~ an atmospheric pressure chemical ionizer.

39. (new) A method of in-process ratio mass spectrometry comprising

- providing a sample having an analyte,
- providing a spike related to the analyte,
- spiking the sample with the spike and permitting equilibrium to occur therebetween,
- in effecting the equilibrium between the spike and the sample, dynamically transforming the analyte and the spike to the same species,
- subjecting the equilibrated spike and sample to atmospheric pressure ionization to create ions therefrom,
- introducing said ions into a mass spectrometer such that the mass spectrometer forms a ratio response, and
- in a processor, using the ratio to characterize the analyte in the sample.

40. (new) The method of in-process ratio mass spectrometry of claim 39, wherein the dynamic transformation of the analyte and the spike to the same species comprises dynamically transforming the spike to the same species as the analyte.

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41 (new) A method of in-process ratio mass spectrometry, comprising
analyzing a first sample from a source having an analyte;
providing a second sample from the source;
providing a spike related to the analyte, the spike having a concentration based upon
the analysis of the first sample;
spiking the second sample with the spike and permitting equilibrium to occur
therebetween,
subjecting the equilibrated spike and second sample to atmospheric pressure ionization
to create ions therefrom,
introducing said ions into a mass spectrometer for a ratio determination, and
in a processor, using the ratio determination to characterize the second sample.

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